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AUTHOR(S):

Kaneyoshi, T.

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Interest in Amorphous Magnetism

T. Kaneyoshi

Department of Physics, Nagoya University, Nagoya 464, Japan

Magnetism in amorphous solids is a relatively new field of physics. Due to the rapid development for over a decade, amorphous magnetism represents already a mature domain of investigation, although a lot of simple questions remain without answer. We have now many reviews and books on the subject. In my talks, three topics are selected from the vast area of research in the field.

1. Critical Phenomena
2. Effects of Transverse Field
3. Ferrimagnetism

1. Critical Phenomena

A crystalline ferromagnet exhibits a sharp phase transition, where many physical properties are divergent. In an amorphous system, on the other hand, there exist both structural and compositional randomness as well as local strains. Therefore, one may expect a smearing of the transition in an amorphous ferromagnet. Experimentally, however, many amorphous ferromagnetic alloys are observed to undergo sharp transitions into their ordered state. They show the normal behavior of a three-dimensional Heisenberg ferromagnet near T_c , with $\gamma=1.38$, where γ is the critical index for magnetic susceptibility; $\chi \propto |T-T_c|^{-\gamma}$ for $T>T_c$. Furthermore, the scaling laws are satisfied within experimental accuracy. This may be expected, if the critical fluctuations very near T_c are of long wavelengths; as long as the system is macroscopically homogeneous, the critical behavior is independent of details of the atomic structure. The critical indices are expected to be the same for macroscopically homogeneous amorphous systems, if the critical exponent of specific heat α continues to be negative even for including the frozen disorder into the ordered system (or the Harris criterion is satisfied). Despite the fact that the critical behavior of more than 50 magnetic glasses has been investigated so far, the effect of frozen disorder on the critical behavior of the ordered system continues to be a mystery. For instance, the specific heat, magnetic and neutron diffraction data on some amorphous ferromagnetic alloys support the Harris criterion, whereas the bulk magnetization, electrical resistivity and Hall effect results on the same or similar glassy ferromagnets refute the Harris criterion.

On the other hand, in amorphous ferromagnetic alloys, there are two features which distinctly reflect the effect of disorder. One is the behavior outside the critical region, the pseudocritical behavior, in macroscopically homogeneous amorphous systems, and the other is the smearing of T_C in strongly disordered systems, which are normally found in weakly itinerant ferromagnets and exhibit an upward curvature for $T > T_C$ in the Arrott plots. In the following, let us discuss the former systems.

In crystalline ferromagnets, the temperature dependences of the inverse paramagnetic susceptibility deviate from the Curie-Weiss law only in the close vicinity of T_C , $|T/T_C - 1| < 0.03$. However, in the amorphous ferromagnets the transition region to the Curie-Weiss law extends over a wide range of temperature, sometimes more than 50% of T_C . This does not necessarily imply that the critical region is wide for disordered systems, but simply means that there is a region of pseudocritical non-linear behavior outside the true critical region. For the systems, a downward curvature of the Arrott plots is always observed in the pseudocritical region. Introducing the Kouvel-Fisher relation defined by

$$\gamma(T) = - \frac{\partial \ln \chi(T)}{\partial \ln (T - T_C)} \quad \text{for } T > T_C,$$

the systems exhibit that $\gamma(T)$ initially increases with temperature and then decreases to the classical value $\gamma=1$, in contrast with crystalline ferromagnets showing a continuous decrease in $\gamma(T)$ from the value at $T=T_C$ ($\gamma=1.34$ for Ni) towards a high-temperature value of unity.

Now, in the above parts we have briefly reviewed the critical and pseudocritical phenomena observed in macroscopically homogeneous amorphous ferromagnets. When we construct a theoretical work, it needs to explain the whole aspects of the phenomena consistently. Two groups have proposed theoretical works, in order to explain the characteristic behavior observed in the pseudocritical region; Stuttgart group (Kronmüller, Fähnle et al.) and Nagoya group (Kaneyoshi et al.). The former uses a self-consistent version of the non-local Landau-Ginzburg theory taking into account the cooperative spin fluctuations and the latter bases on the framework of effective-field theory with correlations. These theories are qualitatively capable of explaining the downward curvature of the Arrott plot isotherms, the upward curvature of χ^{-1} and the compositional dependence of $\gamma(T)$. The background of these theories is due

to the following concept; at $T=T_c$, the thermal spin correlation length ξ is infinite, so that the effects of magnetic inhomogeneity on the physical properties is not felt at all. As the temperature is increased beyond the critical region, ξ decreases. When a temperature is reached, ξ nearly equals to the capillary dimension of structurally induced local magnetic inhomogeneities (such as magnetic clusters). The magnetic inhomogeneity in the system is then no longer averaged out and, as is observed in the pseudocritical region, the characteristic behavior of the Arrott plots, χ^{-1} and $\gamma(T)$ may appear. However, these theories are basically effective-field theories and in many finer details they fail to explain these phenomena. Thus, more elaborate discussions are needed by the use of more sophisticated theories.

Finally, if one is interested in the present experimental status of the critical phenomena in amorphous ferromagnets, Kaul's review paper may be suggested (S.N. Kaul, J. Mag. Mag. Mat. 53 (1985) 5).

2. Effects of Transverse Field

The Ising model with a transverse field was originally introduced by de Gennes as a variable model for hydrogen-bonded ferroelectrics such as the KH_2PO_4 type. Since then, it has been applied to several other systems; for example, cooperative Jahn-Teller systems like DyVO_2 and ferromagnets with strong uniaxial anisotropy in a transverse magnetic field. The model Hamiltonian is given by

$$H = -\sum_i \Omega_i s_i^x - \frac{1}{2} \sum_{ij} J_{ij} s_i^z s_j^z, \quad (1)$$

where s_i^x and s_i^z are spin $-\frac{1}{2}$ operators at site i , Ω_i represents a transverse field, J_{ij} is an exchange interaction, and the sums extend over the points of a lattice.

In two or more dimensions the transverse Ising model has a finite transition temperature, which can be depressed to zero temperature by increasing the transverse field to a critical value Ω_c . An exact solution for the one-dimensional case has been obtained, where no phase transition is verified at finite temperature, but at $T=0$ the system is ordered for Ω less than some critical value Ω_c . Thus, the critical frontier starts at some Ω_c for $T=0$, and ends at the Ising critical point for $\Omega=0$, and it separates the paramagnetic region ($\langle s_i^z \rangle = 0$) from the ferromagnetic one ($\langle s_i^z \rangle \neq 0$) by a second-order phase

transition. At all temperatures, however, there is an order with $\langle s_i^x \rangle \neq 0$.

In the last decade, there has been interest in the problem of disorder in the transverse Ising model, which may apply to KD_2PO_4 - KH_2PO_4 mixed systems and diluted vanadates. The bond- and site-diluted transverse Ising models have been studied by a variety of sophisticated techniques. For the diluted systems, attention has, in particular, been directed to the Harris conjecture that the critical transverse field as a function of concentration at zero temperature should display discontinuity at the percolation concentration.

Now, the magnetism of amorphous solids has been extensively studied. At the present time we do not have any experimental results of amorphous magnets measured in an applied transverse field. Theoretically, therefore, it is interesting to clarify the effects of transverse field on the physical properties in amorphous ferromagnets. On the other hand, because of the difficulties inherent in the theoretical description of amorphous magnets, it is sometimes necessary to make some simplifications. For studying such systems, the lattice model has often been applied, in which the structural disorder is replaced by the random distribution of exchange integral in the Hamiltonian (1), namely

$$P(J_{ij}) = \frac{1}{2}[\delta(J_{ij}-J-\Delta J) + \delta(J-J+\Delta J)] \quad (2)$$

The parameter δ defined by $\delta = \frac{\Delta J}{2J}$ is often introduced in the treatment. The parameter δ is called the structural fluctuation and measures the amount of fluctuation of the exchange interaction coming from the structural disorder in amorphous magnets.

In this part, let us summarize the results of physical properties expected in amorphous ferromagnets, when an transverse field is applied. For the theoretical discussions, see two papers (T. Kaneyoshi, Phys. Rev. B 33 (1986) 526 and J. Phys. Soc. Jpn 54 (1985) 3514). A number of interesting results due to an applied transverse field are found in the thermal behavior. The reduced longitudinal magnetization curve for a small value of Ω falls below that of the corresponding crystalline ferromagnet upon increasing the structural fluctuation, a phenomenon generally observed in amorphous ferromagnets with $\Omega=0$. For a large value of Ω , however, the behavior of the reduced magnetization curve is different from that for a small value of Ω ; the reduced magnetization curve with $\delta=0.35$ is above that of $\delta=0$. On the other hand, for a small value of Ω ,

increasing the value of δ , the saturation value of transverse magnetization m_x increases and m_x becomes extremely sensitive to temperature below the transition temperature, in contrast with that of a crystalline ferromagnet (with $\delta=0$); the magnetization curve decreases rapidly with increasing temperature. For a large value of Ω , the situation of m_x changes; m_x becomes insensitive to temperature.

As is seen from (2), when the value of δ becomes larger than $\delta=0.5$, one of J_{ij} in (2) becomes negative and then it can be expected that the effects of so called frustration may appear in the system. Investigating the phase diagram in the (T_c, δ) space, we find that the possibility of the reentrant phenomenon is preserved for small values of Ω , but it disappears at a value near $\Omega=0.6J$; if the reentrant phenomenon exists in a system with $\Omega=0$, there is a critical transverse field at which the reentrant phenomenon disappears, when we apply an appropriate transverse field to the system.

3. Ferrimagnetism

Amorphous ferrimagnetic rare earth (RE)-transition metal (TM) alloys are currently of considerable interest because of their potential device applications. The alloys have provided an excellent system for investigating basic magnetic phenomena in amorphous magnetic materials. The magnetic properties of the alloys are sensitive to slight compositional changes. The experimental data confirm their untypical magnetic behavior. Of special interest are two physical parameters; the Curie temperature T_c , and the compensation temperature T_{comp} , at which macroscopic magnetizations of both components are identical and cancel each other, being different from zero. Especially, amorphous ferrimagnetic Gd-Co films express compensation points which vary with composition from approximately 40 to 500°K. Compositions of amorphous RE-TM alloys which lead to compensation points in the vicinity of room temperature have been investigated by many authors, after the suggestion of Chavdhari et al. that thermomagnetic writing and erasing can be carried out at $T=T_{comp}$ in these films.

Let us now take the following Hamiltonian

$$H = -\sum_{i < j} \{ J_{AA} \delta_{iA} \delta_{jA} + J_{BB} \delta_{iB} \delta_{jB} + (J_{AB} \delta_{iA} \delta_{jB} + J_{BA} \delta_{iB} \delta_{jA}) \} s_i^z s_j^z \quad (3)$$

where the A- and B- atoms have different spins ($s_A = \frac{1}{2}$ and $s_B = 1$) respectively and

the sum is over all nearest-neighbour pairs. ξ_i is a random variable which takes the value of unity or zero, depending on whether the site i is occupied by a magnetic atom or not. In real systems J_{AA} , J_{BA} and J_{BB} correspond to TM-TM, RE-TM and RE-RE interactions. The magnitudes of exchange interactions are usually taken as $J_{BB} < -J_{BA} < J_{AA}$.

In order to analyze the experimental data of amorphous RE-TM ferrimagnetic alloys, all experimentalists have taken a model; RE and TM ions are randomly distributed in a lattice with $z=12$ and the exchange interaction between two ions is determined entirely by the species of those ions. The sublattice magnetizations m_A and m_B are then assumed to follow Brillouin functions (or the MFA theory) for the spin values s_{RE} and s_{TM} ($s_{TM} < s_{RE}$).

From a great number of experimental works, in amorphous TM-metalloid alloys the fluctuation of exchange interaction J_{AA} is considered as an important ingredient for the appearance of characteristic behavior, such as the depression of reduced magnetization curves, reentrant phenomena and spin-glass phase. In amorphous Gd-noble metal ferromagnetic alloys, on the other hand, reentrant phenomena and spin-glass phase are also found; the exchange interaction J_{BB} is considered to be fluctuating around a mean value. Thus, in amorphous RE-TM ferrimagnetic alloys, it may be reasonable to take account of the structural fluctuation, like (2), although it is not sure whether the exchange interaction J_{AB} fluctuates around a mean value.

To our knowledge, however, the effects of the structural fluctuation δ ($\delta = \frac{\Delta J_{AA}}{J_{AA}} = \frac{2\Delta J_{AB}}{J_{AA}} = \frac{2\Delta J_{BB}}{J_{AA}}$) on T_C , T_{comp} and sublattice magnetizations in amorphous ferrimagnets have not been discussed in previous work. Theoretically, when we use the MFA (mean-field approximation) theory, it is known that the effect of δ on T_C cancels out and does not appear, and its effect on T_{comp} is very small. The results, however, are characteristic to the MFA. In this way, it is necessary to investigate the effects of the structural fluctuation on the magnetic properties within the framework over the standard MFA theory.

Using the effective-field theory with correlations introduced by us (for $\delta=0$ it corresponds to the Zernike approximation), we have studied the effects of δ on the magnetic properties. A number of interesting effects of δ comes up in the behavior of T_C and T_{comp} ; in general, the effect of δ on T_C and T_{comp} is to fall below the values of T_C and T_{comp} in the disordered ferrimagnetic crystalline alloy, which phenomenon is observed in real amorphous ferrimagnetic alloys. On the other hand, when one of exchange interactions becomes to take

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positive and negative values randomly, the so-called frustration effect appears in the T_c , T_{comp} and sublattice magnetizations. The results imply that the MFA theory not including the effect of structural fluctuation may give incorrect analyses of the experimental data for T_c and T_{comp} and the MFA theory must be applied with caution to real amorphous ferrimagnetic alloys. Moreover, our results indicate the possibility of reentrant ferrimagnetic alloys.

Finally, at the present time, the main interest of experimentalists is directed to obtain an amorphous ferrimagnetic alloy which has a compensation point in the vicinity of room temperature, because of its potential device applications, such as the thermomagnetic recording. Our results propose that the research of a new amorphous ferrimagnetic alloy may have the possibility of new phenomena coming from the random distribution of exchange bonds.